A tribute to Robin Hochstrasser

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A Tribute to Robin Hochstrasser

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This Special Issue was originally planed to honor Robin Hochstrasser's long-lasting commitment as Editor of Chemical Physics. Events took a sad turn when we heard of Robin's passing. The Special Issue is a collection of research papers from former co-workers of Robin, as well as from close friends and colleagues. As such, this Special Issue documents in a sadly timely fashion the tremendous impact Robin has had on the community and the breadth of his research. In particular the latter is reflected in the wide range of research fields from his former co-workers. Starting their career in Robin's lab, they went on to develop their own research with the inspiration they received from Robin. The large number of people that had the privilege to interact with Robin, many of whom contribute to this Special Issue, guarantees that his spirit and his view of science will live on. As briefly discussed below, the collection of papers in this Special Issue gives the reader an idea to what extent this statement is correct.

Robin influenced and led not just one, but several generations of physical chemists and biophysicists. By striking out into the new field of ultrafast spectroscopy when it was just being born, Robin changed the nature of spectroscopy. Robin realized well before others that moving to the picosecond and shorter timescales was not just about measuring faster and faster events, but was about revealing and understanding the fundamental processes that underlay all condensed phase chemical and biochemical phenomena. Robin went on to apply his techniques, especially those involving infrared ultrashort pulses to biophysical problems, especially

peptide and protein structure and dynamics, this, at a time when it was far from fashionable for 'real' physical chemists to study biological systems. But Robin did not follow fashion or fads, rather his own insatiable curiosity and intellectual agenda. A glance at Robin's publications [1–505] reveals the astonishing range of topics and the consistency in setting new benchmarks.

Over the last 15 years, Robin's research strongly focused on 2D IR spectroscopy and its applications to study structure and ultrafast dynamics of all sorts of molecular systems, ranging from small molecules to proteins, with unprecedented detail. His groups was the first to introduce that technique in 1998 [117], and it underwent a tremendous development since then. From a methodology side, the development is documented in the paper by Martin Zanni, who spends significant effort to simplify the technique, up to the point that he is currently commercializing it in order to make 2D IR spectroscopy available to a wider community. During these 15 years, quite a number of students and postdocs were trained in Robin's lab and went on to become successful professors all around the world, continuing to develop 2D IR spectroscopy. Some of them contributed to this Special Issue, for example Igor Rubtsov, who uses a variant of 2D spectroscopy, relaxation assisted 2D IR spectroscopy, to measure vibrational energy transport in molecular chains, and Nien-Hui Ge, who applies dual frequency 2D-IR spectroscopy to elucidate correlation between two modes in ethyl diazoacetate.

2D spectroscopy covering the THz to the UV spectral range was picked up by many other research groups worldwide, not directly connected to Robin, as a promising new technique. An estimated number of 50-70 research groups now pursue 2D optical spectroscopy. Examples are found in the paper of Charles B. Harris, a long-standing friend of Robin, who studies the mass effect on rotational diffusion of small solutes in solution, and in the paper of Minhaeng Cho, which reports on the ultrafast intermolecular vibrational excitation transfer from a solute into the solvent. Majed Chergui, a fellow editor of Chemical Physics, now pushes 2D

spectroscopy into the UV range, and lays the foundations of 2D UV spectroscopy of tryptophan in his paper. From a theoretical point of view, 2D spectroscopy opened many new avenues. For example, Wolfgang Domcke, another fellow editor of Chemical Physics, discusses the calculation of third-order optical signals via a driven Schrödinger equation with applications for 2D spectroscopy, and Shaul Mukamel simulates the 2D IR and stimulated resonance Raman spectra of the photoactive yellow protein.

Quite recently, Robin applied 2D IR spectroscopy to study the structure of the influenza A channel [10], to that of amyloid fibrils [25], as well as the dynamics of water confined in a protein environment [18]. Using more conventional spectroscopic tools, similar problems are addressed in the papers by William F DeGrado, a close collaborator of Robin, and by Sudipta Maiti and Joël Friedman, both former co-workers of Robin.

Time-resolved vibrational spectroscopy on biological systems played an important role in Robin's science even before the invention of 2D IR spectroscopy. A landmark paper in this regard as early as 1989 showed by femtosecond transient IR spectroscopy of the photon induced disappearance of the Fe-CO bond in hemoglobin that the bond breaking is faster than 200 fs [215]. The first author of that paper, Philip Anfinrud, continues to work on related questions, now with very elegant time resolved x-ray diffraction experiments. Furthermore, Manho Lim, a postdoc with Robin, investigates the vibrational relaxation of NO stretching modes in ferrous NO and ferric NO in a model heme.

Robin was always intrigued by the chemical selectivity of vibrational spectroscopy. In combination with the intrinsic high time-resolution , this allows one to investigate ultrafast structural processes in great detail. Such experiments require a fast trigger to initiate a structural transition. In 1997, Robin was the first to introduce the idea of using a photochemical reaction, in this case the light-triggered photodissociation of a disulfide bridge, as a trigger to initiate α-helix folding [125]. In their papers, Martin

Volk and Peter Hamm, both former postdocs with Robin, follow up on that idea and use ultrafast temperature or pH-jumps, respectively, to induce the folding of model α-helical peptide polyglutamic acid. The response of the peptide is monitored by transient IR spectroscopy. Similarly, his next-door colleague Feng Gai uses temperature jumps in connection with D-amino acids to delineate the folding mechanism of the Trp-cage.

To understand vibrational energy flow on a more fundamental level, Robin studied the ultrafast vibrational energy relaxation in small molecules and ions, work which has had lasting influence on the experimental and theoretical growth of this field [196,299]. Jeffrey Owrutsky and Ted Heilweil were the PhD students responsible for that work, and report on a related problem in their paper.

Robin always combined experimental virtuosity with deep theoretical analysis. One of the intriguing aspects of all of the experiments described so far is the fact that the timescale of the investigated phenomena and size of the molecular system very well match what can be reached with molecular dynamics (MD) simulations, which in turn can provide a very detailed atomistic picture and thus much insight into the processes. Gnana Gnanakaran was a PhD student of Robin working on simulations, and he continues to be successful with molecular dynamics work, as documented in his paper, where he discusses how Bayesian statistics can deduce the conformational variability of intrinsically disordered proteins from infrared spectroscopy. Many of Robin's theory colleagues at University of Pennsylvania are represented as well. Michael Klein determines the proton affinity of the histidine-tryptophan cluster motif from the influenza A virus from ab initio molecular dynamics, Gregory Voth develops coarse-grained models of proteins based on elastic network, and Andrew Rappe studies the picosecond phenol migration in phenylacetylene solution.

While most of the more recent work of Robin emphasized the

dynamics of molecular systems on electronic ground state potential energy surface, there are, of course, also important biological processes that involve electronically excited states. The photodissociation of Fe-CO bond in haemoglobin [215] or the photoisomerization of retinal in bacteriorhodopsin [148] are just two examples from Robin's lab. Many of his former co-workers study a wide spread of similar processes, such as the paper of Angus John Bain on the activated barrier crossing dynamics in the nonradiative decay of NADH and NADPH, that of Bernhard Dick on the electronic spectroscopy to lumiflavin in superfluid helium nanodroplets, that of Chong Fang on the excited-state proton transfer of photoexcited pyranine in water by femtosecond stimulated Raman spectroscopy, and that of Roseanne Sension on the use of broadband ultrafast transient absorption to investigate Fe(III)TPPCl in the condensed phase.

In the late 90's, Robin also developed a strong research line on single molecule spectroscopy, next to the ultrafast work that dominated the group'activity at that time. He was the first to measure by single-molecule confocal microscopy the folding and unfolding conformational equilibrium distributions and dynamics of protein immobilized on a surface [97]. Along the same lines, Bill Eaton, former PhD student and close friend of Robin, measures ultrafast protein folding rates from photon-by-photon analysis of single molecule fluorescence trajectory. Carey Johnson, former postdoc with Robin, analyses single molecule FRET burst measurements by Bayesian nested sampling.

In 1985, Robin, together with graduate student Minyung Lee, was one of the first to observe the long sought-after "Kramers turnover" in the isomerization rate constant as a function of pressure, density and viscosity using supercritical ethane as solvent [262]. In his paper, Minyung Lee again works with photoisomerizing dyes, now as tool to determine the elastic modulus of poly(ethylene oxide).

In the 60's and 70's, Robins research focused on the spectroscopy

of molecular crystals, where the coupling between molecules leads to excitonic states, a phenomenon which had seen a revival in his work much later when studying the amide I of peptides and proteins [117]. An important topic at that time were ultrahigh magnetic field Zeeman effects in triplet states and the electric field induced Stark effects on molecular excitons [447]. Tom Lin, co-author in that paper, continued along these lines and discusses the dynamics of nuclear polarization by optical pumping in zero-field. Already in these early days, Robin realized that the dimensionality of the molecular crystal has dramatic effect on the spectroscopy [412]. This idea forms the basis of the very active field of nanomaterials spectroscopy. Many of Robin's former co-workers are extremely active in this area now. For example, Bruce Weisman, discusses the length-dependent optical properties of single-walled carbon nanotube samples, Tim Lian, his successor as editor for Chemical Physics, the vibrational relaxation dynamics of catalysts on rutile TiO_2 (110) surfaces, Anne Myers Kelley the resonance Raman spectra of wurtzite and zincblende CdSe nanocrystals, and Barry Dellinger the formation of environmentally persistent free radicals from phenol adsorption on Al_2O_3 and TiO_2.

It is hard to find a topic of Physical Chemistry in which Robin has not been very active. But there are in fact two papers in this Special Issue that deserve that label, namely the paper by Kent Blasie on the structural changes in single membranes in response to an applied transmembrane electric potential revealed by time-resolved neutron and X-ray interferometry, and that of Hai-Lung Dai on the collision-induced intersystem crossing in the ketenyl (HCCO) radical. Both authors were colleagues of Robin at the University of Pennsylvania.

The extraordinary progress in our understanding of condensed phase structure and dynamics over the past 50 years is scarcely conceivable without the massive contributions of Robin Hochstrasser. This volume is a small tribute to that legacy.

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